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Sulfate and Nitrate Collected by Filter Sampling Near the Tropopause

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SUMMARY

Filter samples collected in the lower stratosphere and upper troposphere during 1976, 1977, and 1978 with a NASA Lewis F-106 aircraft and two commercial Boeing 747 aircraft were analyzed for sulfate and nitrate ions. These sampling platforms were operated as part of the NASA Global Atmospheric Sampling Program (GASP). The sulfate mixing ratios were generally highest just above the tropopause. The nitrate mixing ratios were generally more variable than the sulfate mixing ratios.

Average sulfate and nitrate mixing ratios were generally high in the winter-spring period, with a moderate peak in the stratosphere just above the tropopause. Mixing ratios were substantially lower in the summer-fall period, with no significant differences between tropospheric and stratospheric samples near the tropopause.

Combining the 747 data with the F-106 data in chronological order for commercial flight altitudes at or below 12.5 kilometers revealed similarities and overall characteristics. In this altitude range, stratospheric mass mixing ratios averaged 0.26 ppbm for sulfate and 0.35 ppbm for nitrate for the winter-spring group and 0.13 ppbm for sulfate and 0.25 ppbm for nitrate for the summer-fall group. Tropospheric mass mixing ratios averaged 0.08 ppbm for sulfate and 0.10 ppbm for nitrate for the winter-spring group and 0.05 ppbm for sulfate and 0.08 ppbm for nitrate for the summer-fall group.

Sulfate mixing ratios correlated well with nitrate mixing ratios, with a correlation coefficient of 0.78. Ozone data also correlated well with the sulfate and nitrate mixing ratios, with correlation coefficients of 0.80 and 0.79, respectively. These correlations are evidence of the transport of sulfate and nitrate ions from the stratosphere to the troposphere.

INTRODUCTION

In this investigation the sulfate ($SO_4^{=}$) and nitrate (NO_3^{-}) ion content of aircraft filter samples collected near the tropopause during 1976, 1977, and 1978 was determined. The data supplement the measurements reported by other investigators (refs. 1 to 4) by extending the altitude range from the peak sulfate region, located above 14 kilometers downward to the boundary region near the tropopause.

Several recent publications (refs. 5 to 8) have expressed concern about the effects of aircraft emissions on the environment. One-dimensional kinetic calculations have shown that gas-phase reactions can produce a layer of sulfate directly above the tropopause (refs. 9 and 10). In addition, concentrations of nitric acid vapor, determined by analysis of balloon and aircraft filter samples, show a broad peak above 20 kilometers (refs. 11 and 12).

NASA began the Global Atmospheric Sampling Program (GASP) several years ago to develop an automated system for obtaining world-wide measurements of atmospheric constituents that may be affected by aircraft exhaust emissions (ref. 13). Aerosols and their precursors and oxides of nitrogen and their acid products seemed to be of particular interest for studying formation and transport processes and for developing atmospheric models.

As part of GASP, filter sampling systems were designed for use on a NASA Lewis F-106 aircraft for local and special sampling and on two commercial Boeing 747 aircraft for automated routine sampling (ref. 14). On the F-106 the filter sampling system was installed in a pod mounted under each wing. On the 747's the filter sampling system was installed within the fuselage below the passenger level near the front of the aircraft. Sampling altitudes were from 8.8 to 13.7 kilometers. Routine filter-sample exposure time was 90 minutes on the F-106 and 120 minutes on the 747. The filter collection medium was primarily IPC-1478 material, which is commonly used for high-altitude air sampling.

Filter analysis techniques included atomic absorption spectroscopy, electrochemistry (specific-ion electrodes), and ion chromatography. Only the filter analysis results for sulfate and nitrate concentrations are presented. Other anions identified, namely Cl^- and F^-, generally were at a very low collection level that was within the range of background variability.

APPARATUS AND PROCEDURE

Sampling Systems

Single- and multifilter sampling systems were designed and developed by NASA for the GASP aircraft. The single-filter system was originally designed for use on the F-106; however, two single-filter systems were also adapted for interim use on the 747's until the multifilter system became available.

The single-filter system is shown in figure 1: an internal view of the mechanism in figure 1(a), and the filter-holder assembly in figure 1(b). The system contains a single filter holder that is inserted into a sampling duct for filter collection and then is retracted and stored on command by electrical control. A filter system was installed in a pod under each wing of the F-106, as typically shown in figure 2. The F-106 pod assembly consists of a 25-millimeter-diameter inlet tube that expands to a 76-millimeter-diameter duct for sample collection. The filter mechanism and the inlet and exit valves are controlled from the cockpit by the pilot. Airflow measurements were determined from venturi flow data printed out on an auxiliary recorder at 1-minute intervals. Supplemental data--including valve positions, mechanism status, sample air temperatures, and system pressures--were also recorded.

So that a preliminary evaluation could be made of the automatic filter sampling capability of the GASP system, two single-filter systems were adapted for use on two 747 aircraft, as shown in figure 3. Modifications included an easy-access door, an auxiliary control box with filter pack identification switches, and mounting brackets. The single-filter system installed on the 747 is shown in figure 4.

The multifilter system is shown in figure 5: an internal view of the mechanism in figure 5(a), the filter-holder assembly in figure 5(b), and the magazine assembly in figure 5(c). The multifilter system installed on the 747 is shown in figure 6.

The multifilter system is an enclosed slide mechanism that accommodates a filter magazine containing eight individual filter holders protected from contamination by close-fitting separator plates positioned between adjacent holders. Filters are inserted, retracted, and advanced automatically on command from the GASP system control. Filters are identified by a dialed-in pack number for each magazine and by cam-actuated filter sequence identification switches for individual filter numbering. The first and seventh filters in a magazine are normally reference blanks that are briefly inserted into the sampling position for 1 minute during routine sequential cycling. When the reference filter (which is not exposed to airflow) is retracted, the next filter is immediately inserted and

exposed during the same flight. Airflow for the filter system is supplied from an air inlet probe (25-mm diam). System cleanliness is maintained by capping the inlet probe during takeoff and landing. To prevent airflow during filter insertion or retraction, the GASP control system automatically closes a filter vent valve. A venturi flowmeter located downstream of the filter is used to measure airflow rates. The 747 sampling system is described further in references 14 and 15. Both single- and multifilter systems operated satisfactorily in service with no discernible change in filter sample integrity.

Filter Preparation

The filter collection medium was primarily IPC-1478. This is a low-resistance, cellulose material made from second-cut cotton linters with cotton scrim backing for added strength. This paper was specially designed by the Institute of Paper Chemistry (ref. 16) for high-altitude air sampling and thus features low pressure drop, low flow resistance, and good retention of small airborne particles. It is impregnated with Kronisol (dibutoxyethyl phthalate) during manufacture to improve its collection efficiency.

An alternative filter collection medium, Gelman Type A glass-fiber filter material, was used on several flights in conjunction with the IPC-1478 material, and the collection results were compared. The glass-fiber filters have moderate flow resistance.

Before use, all filters must be washed to remove residual amounts of water-soluble contaminants (ref. 17). A semi-automatic apparatus (fig. 7) is available that washes 25 filters at a time. A tray is loaded with filters, each sandwiched between stainless-steel support screens. The washing procedure is as follows:

(1) Immerse a tray of filters in carbonate buffer solution (0.024 M sodium carbonate and 0.030 M sodium bicarbonate) and soak for 5 to 10 minutes.

(2) Rinse three times in deionized water.

(3) Immerse in 0.1 M acetic acid solution and soak for 3 to 5 minutes.

(4) Rinse three times in deionized water.

(5) Wash IPC-1478 filter trays at least six times in automatic cycling washer system with deionized water saturated with dibutoxyethyl phthalate. Wash Gelman Type A filters individually with plain deionized water. Use a coarse, fritted disk funnel to support individual filters.

(6) Dry in desiccator over potassium hydroxide (KOH) pellets at 0.20 N/cm^2 (15 mm Hg) for at least 24 hours.

All chemicals are reagent grade. All procedures are conducted at room temperature.

Samples from each wash group were analyzed for background levels of contamination in order to verify the washing procedure. Washed filters with residual sulfate levels to 2 micrograms per filter and residual nitrate levels to 1 microgram per filter were acceptable. Upon acceptance, the filters were transferred to a clean room for assembly into filter holders. Contamination during shipping and handling was prevented by always sealing single-filter holders or multifilter magazines before and after use in ultraclean polyethylene bags conforming to cleanliness levels of NASA specification KSC-C-1230.

Filter Sampling

The F-106 sampling altitude ranged from 8.8 to 13.7 kilometers and was usually a preselected altitude increment from the reported tropopause location. The 747 sampling altitude was limited by normal commercial flight operations to 9.6 to 12.5 kilometers. Routine filter exposure time on the F-106 was 90 minutes (controlled manually by the

pilot), with occasional variations depending on flight altitude, weather conditions, and flight plan. Routine filter exposure time on the 747 was 120 minutes unless sampling was automatically stopped because the aircraft had descended below 9.6 kilometers.

The paired filter sampling capability of the F-106 aircraft was used on some flights to compare alternative filter materials such as a Gelman Type A glass fiber or special chemically treated IPC-1478 filters. Most flights, however, were used for dual simultaneous sampling with the standard IPC-1478 material.

For typical 747 flight conditions (flight Mach number of 0.82), the airflow rate through IPC-1478 filter material was nominally 3.60 ambient m^3/min at 10.7 kilometers altitude. For typical F-106 flight conditions (flight Mach number of 0.88), the airflow rate through IPC-1478 was nominally 3.88 ambient m^3/min at 11.3 kilometers altitude. These flow rates correspond to choked flow in the venturi and to essentially constant volumetric flow at these sampling altitudes and flight conditions. Although these flow rates do not give isokinetic sampling at the inlet probe, it is generally believed that the upper atmosphere aerosol consists predominantly of submicrometer-size particles that should follow the gas streamlines.

The airflow rates through the glass-fiber filter material at a flight Mach number of 0.88 were 1.26, 1.51, and 1.76 ambient m^3/min at altitudes of 13.7, 13.1, and 11.3 kilometers, respectively. With the glass-fiber material the venturi does not choke at these flight conditions, and thus the airflow rate varies with flight altitude.

The collection efficiency of IPC-1478 filter paper at normal atmospheric pressure as a function of particle size, filter face velocity, and filter mass loading are discussed in reference 18. Typical filter face velocities for IPC-1478 were 800 m/min for the 747 flight conditions and about 850 m/min for the F-106 flight conditions. The impregnated IPC-1478 was more than 90 percent efficient at face velocities of 600 m/min with 0.312-, 0.557-, and 1.011-micrometer-diameter latex particle spheres. The efficiency increased with increasing face velocity for particles under 0.3 micrometer diameter; however, for large particles the efficiency was highest at lower velocities. Thus, at very low loading conditions, such as at sampling altitudes near the tropopause, the impregnated IPC-1478 has a relatively high collection efficiency at the velocities and conditions encountered with the GASP aircraft.

Filter Analysis

Before analysis, each filter is cut into four equal segments so that segments can be reserved for separate constituent analysis, for comparative repeat analyses, and for future use. Originally, both spectrophotometric and electrochemical techniques were used to analyze for each anion of interest separately. Thus, sulfate ion concentration was determined indirectly by flame emission spectroscopy, that is, by measuring the amount of barium ion that remained after a known addition of barium ion in a solution reacted with the filter segment. Nitrate and chloride ion concentrations were determined by using specific-ion electrodes to measure the specific-ion potential in a solution produced by wetting a filter segment in an extracting solution of sulfuric acid. These techniques are described further in reference 19.

Ion chromatography was used for all filter analysis after about mid-1976. Comparative analyses showed no gross differences between this technique and those used previously. Ion chromatography was preferred over the other techniques because all anions of interest ($SO_4^=$, NO_3^-, Cl^-, and F^-) are obtained on one chromatogram and thus the method is faster. Other advantages are greater sensitivity and precision and less influence from interferring substances.

The basics of ion chromatography are described in references 20 to 22. This procedure requires wetting a filter segment with an extracting solution of 10 milliliters of carbonate buffer (0.0024 M sodium carbonate and 0.003 M sodium bicarbonate). A 0.5-milliliter sample is injected into the ion chromatograph flow system, which includes a carbonate eluant background stream, an anion separator column, a suppressor column for anion conversion to its acid form, and a conductivity detector. The instrument is calibrated by using solutions with known concentrations of the various anions in the extractant. The anion concentration is calculated by comparing the constituent peak heights from the sample chromatograms with those obtained with the standard calibrating solution.

The net amount of any constituent on a filter is calculated by subtracting an average background level determined from several reference filters reserved from each group of filter assemblies or obtained from reference cycle insertions. Ion chromatographic analysis of reference filters (from both F-106 and 747 sampling) yielded group background sulfate levels averaging 2.4 micrograms per filter (124 samples) and group background nitrate levels averaging 6.8 micrograms per filter (122 samples). The standard deviation in the filter background levels amounted to $\pm$1.6 micrograms per filter for sulfate and $\pm$3.1 micrograms per filter for nitrate. Thus, there is an uncertainty in the constituent measurements because of the variability in the background levels. For comparison, a variability of $\pm$1.0 microgram in the background level represents an uncertainty of 0.010 ppbm per 100 000 grams of sample air.

Sampling at 747 flight conditions at an altitude of 10.7 kilometers for 120 minutes resulted in uncertainties of 0.0098 ppbm for sulfate and 0.019 ppbm for nitrate. Proportionately greater uncertainty is associated with shorter filter exposure times and higher sampling altitudes, for which the total air mass sampled is reduced. Thus, sampling at F-106 flight conditions at an altitude of 13.7 kilometers for 90 minutes resulted in uncertainties of 0.019 ppbm for sulfate and 0.037 ppbm for nitrate.

The variability in the filter background levels when the spectrophotometric and electrochemical analysis techniques were used was about $\pm$6 micrograms for sulfate and $\pm$4 micrograms for nitrate. Thus, significantly greater uncertainty was associated with using the original analysis techniques.

RESULTS AND DISCUSSION

Sulfate ($SO_4^=$) and nitrate (NO_3^-) data obtained during 1976, 1977, and 1978 from analysis of aircraft filter samples collected at altitudes near the tropopause are listed in tables I to IV. The data presented in tableII were obtained with the NASA Lewis F-106 aircraft and are arranged by seasonal flight series. The F-106 data flights originated from Cleveland, except for those during February 1976 (Holloman AFB, N. Mex.), October 1976 (Ellington AFB, Tex.) and May 1977 (Elmendorf AFB, Alaska). The data presented in table II were obtained with single-filter sampling systems installed on the two GASP Boeing 747 aircraft during regularly scheduled flights by United Airlines and/or Pan American Airlines (see refs. 19, 23, 24, and 25 for availability of supplemental data). The data presented in table II were obtained with multifilter sampling systems installed on the same 747 aircraft during flights in 1977 and 1978 (see ref. 25 for availability of supplemental data). The data presented in table IV provide a statistical summary for comparing seasonal and altitude variations. From all these data, various plots are presented to show seasonal, altitude, and chronological characteristics; accompanying correlations show corresponding data relationships.

Altitude profiles of the sulfate mixing ratio data from table I are presented in figures 8 and 9 for the winter-spring and summer-fall, respectively, of 1976 and 1977. The corresponding nitrate mixing ratio data from table I are similarly presented in figures 10 and 11, respectively. The data are plotted at differential altitude pressure intervals from the tropopause to show the altitude characteristics and seasonal variations.

Generally, sulfate and nitrate mixing ratios were much higher in the winter-spring than in the summer-fall. The sulfate mixing ratios generally were highest just above the tropopause. The nitrate data generally were more variable than the sulfate data. Sampling between 30° and 70° N latitude did not reveal significant differences in the altitude or seasonal characteristics from those observed at Cleveland.

Mixing ratio data from the F-106 samples averaged for the winter-spring and the summer-fall groups are shown in table IV (group A). Stratospheric mass mixing ratios averaged 0.35 ppbm for sulfate and 0.49 ppbm for nitrate for the winter-spring group and 0.11 for sulfate and 0.26 ppbm for nitrate for the summer-fall group. Tropospheric mass mixing ratios averaged 0.16 ppbm for sulfate and 0.11 ppbm for nitrate for the winter-spring group and 0.09 ppbm for sulfate and 0.18 ppbm for nitrate for the summer-fall group.

Data presented in references 2, 4, 11, 12, and 26 give evidence of nitric acid vapor in the stratosphere as the source of the nitrate ion captured on the IPC-1478 filters. Comparative sampling with IPC-1478 and polystyrene filters (ref. 11) demonstrate that nitrate collected on IPC-1478 filters apparently by gas adsorption but that the polystyrene filters did not adsorb nitric acid vapor.

For a further comparison of nitrate collection data, three F-106 flights were made with glass-fiber filter material in one pod and IPC-1478 filter material in the other pod. The resultant sulfate and nitrate data for paired samples shown in figures 9(d) and 11(d) show similar or slightly lower collection levels for the glass-fiber material.

Subsequent laboratory experiments were conducted to verify that nitric acid can be adsorbed on glass-fiber filters. Purified dry air at atmospheric pressure was passed over a 2 to 2.5 M nitric acid solution and through the glass-fiber filters at a flow rate of 1000 cm^3/min. An apparent saturation level of about 6 micrograms nitrate per filter resulted after 5 to 10 minutes of sampling. After initial analysis, the remaining filter segments were stored for about 33 days and then exposed to vacuum at simulated altitude pressures for up to 26 hours. No significant loss of nitrate content resulted.

Since the net collection of nitrate on the F-106 glass-fiber filters was only 2.4 to 8.1 micrograms per filter, the observed nitrate collection probably includes gaseous nitric acid adsorption and retention on the glass-fiber surfaces. Paired samples with much larger collections of nitrate would be required to determine the percentage of particulate nitrate, if any, in the sample. Other studies involving the collection of particulate nitrates on filter materials are discussed in reference 27.

To further demonstrate the general characteristics observed in the separate seasonal flight series, the data were paired by season, averaged in altitude increments of 20 millibars, and replotted as shown in figure 12. Sulfate and nitrate levels are, as expected, higher in the winter-spring periods than in the summer-fall periods. The greater variability in the nitrate data is still apparent. The existence of a secondary sulfate layer, as sug- gested in references 28 and 29, is partially substantiated by the sulfate data; however, ad- ditional data would be desirable.

The 747 filter data (tables II and III) and selected F-106 filter data (table I) obtained at or below 12.5 kilometers altitude are presented in chronological order in figure 13 in order to show overall trends and characteristics for the altitude range routinely sampled by the 747. The sulfate and nitrate data are each further separated as to stratospheric and/or uncertain air samples for comparison with tropospheric air samples. For all the data obtained at and below 12.5 kilometers (group C, table IV), stratospheric mass mixing ratios

averaged 0.26 ppbm for sulfate and 0.35 ppbm for nitrate for the winter-spring group and 0.13 ppbm for sulfate and 0.25 for nitrate for the summer-fall group. Tropospheric mass mixing ratios averaged 0.08 ppbm for sulfate and 0.10 ppbm for nitrate for the winter-spring group and 0.05 ppbm for sulfate and 0.08 ppbm for nitrate for the summer-fall group. Table IV (groups B and D) shows separate averages of 747 data only and all data combined.

Included in table IV are the standard deviations within each data group, which demonstrate the high degree of variability in the measurements. In some cases, the standard deviation was equivalent to the mean of the measurements. Therefore, seasonal differences were not as obvious as differences between exposure regions, especially in the troposphere.

The relationship of nitrate mixing ratios to sulfate mixing ratios is demonstrated by correlating all the data, as shown in figure 14. Linear regression of the data yields a correlation coefficient of 0.78, which is within the range of stratospheric data presented in reference 12. The high correlation of nitrate and sulfate in the vicinity of the tropopause adds further evidence to the reasonableness of the assumption in reference 12 - that both nitrate and sulfate are related through dynamic processes that transport aerosol and nitric acid vapor from a source region in the stratosphere to the troposphere.

The relationship of sulfate and nitrate mixing ratios to ozone mixing ratios is strengthened by separate correlations of the 747 data, as shown in figures 15 and 16, respectively. The high correlation coefficients (0.80, 0.79) of both sulfate and nitrate to ozone support the reasoning for aerosol transport from the stratosphere to the troposphere.

CONCLUDING REMARKS

The filter data obtained during the Global Atmospheric Sampling Program (GASP) show substantial mixing ratio variability, especially for nitrate, and generally exhibit moderately high sulfate concentrations just above the tropopause during the winter-spring. It appears that the natural descent of the tropopause to lower altitudes during the winter-spring period enhances the transport of ozone, sulfate aerosol products, and nitric acid vapor from the stratosphere downward toward the tropopause. The data presented may be useful to characterize the altitude region near the tropopause in future modeling studies. Further filter sampling may be desirable to discern annual trends and changes in mixing ratios.

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TABLE I. - FILTER DATA COLLECTED BY NASA F-106 AIRCRAFT

Flight[a]	Exposure date	Flight altitude, km	Exposure time, min	Interval below or above average tropopause, millibars	$SO_4^=$ concentration, $\mu g/m^3$	$SO_4^=$ mixing ratio, ppbm	NO_3^- concentration, $\mu g/m^3$	NO_3^- mixing ratio, ppbm	Average latitude, deg	Average longitude, deg
[b]32	2-12-76	11.9	90	56	[c]0.049	[c]0.154	[d]0.034	[d]0.107	38N	98W
[b]33	2-12-76	11.9	85	-38	[c].023	[c].072	[d].020	[d].063	35N	104W
34	2-13-76	12.5	85	-35	[c].036	[c].125	[d].000	[d].000	33N	106W
35	2-14-76	13.7	60	0	[c].005	[c].021	[d].037	[d].156	↓	↓
38	2-16-76	13.7	85	108	[c].042	[c].177	[d].042	[d].177		
39	2-16-76	12.5	90	77	[c].073	[c].254	[d].052	[d].181		
40	2-17-76	11.3	90	5	[c].199	[c].571	[d].131	[d].376		
[b]41	2-19-76	12.5	90	33	[c].052	[c].180	[d].102	[d].354	38N	94W
42	5-24-76	13.1	90	40	.110	.422	.133	.510	43N	84W
43	5-24-76	12.5	90	24	.130	.456	.122	.425	↓	↓
44	5-25-76	13.7	75	117	.093	.392	.111	.468		
45	5-26-76	11.9	105	18	.121	.382	.110	.348		
46	5-27-76	13.7	90	41	.124	.522	.109	.460		
47	5-28-76	12.5	105	26	.078	.271	.122	.424		
48	6-2-76	13.1	105	13	.135	.517	.188	.720		
50	7-22-76	12.5	90	-47	.032	.111	.077	.268	42N	86W
51	7-22-76	13.1	↓	-30	.030	.115	.035	.134	42N	↓
52	7-26-76	11.9		-60	.034	.107	.044	.139	40N	
53	7-26-76	12.5		-42	.021	.073	.017	.059	40N	
[b]54	10-26-76	12.5	60	3	.044	.152	.094	.326	35N	90W
55	10-26-76	13.7	70	[e]2	.031	.131	.075	.316	33N	95W
56	10-27-76	13.1	75	-35	.024	.092	.056	.214	33N	96W
58	10-28-76	13.7	75	[e]-7	.025	.105	.046	.194	33N	96W
59	10-28-76	13.7	60	[e]-9	.024	.101	.020	.084	32N	97W
60	10-30-76	13.7	75	[e]-20	.023	.097	.034	.143	31N	99W
61	10-30-76	13.7	77	-33	.019	.080	.032	.135	31N	99W
62	10-31-76	11.9	90	0	.022	.070	.047	.148	30N	93W
[b]63	10-31-76	12.5	70	45	.042	.146	.072	.250	35N	90W
64	3-2-77	13.7	90	102	.106	.447	.199	.840	42N	87W
65	3-3-77	↓	80	78	.064	.270	.150	.633	↓	↓
66	3-8-77		90	75	.113	.477	.161	.679		
67	3-8-77		↓	60	.106	.447	.219	.924		
68	3-9-77	12.2		42	.123	.408	.233	.773	46N	92W
69	3-9-77	12.5		42	.124	.431	.201	.699	46N	92W
70	3-10-77	13.7		69	.083	.350	.132	.556	42N	87W
71	3-15-77	13.7		97	.066	.278	.041	.173	45N	86W
72	3-15-77	13.7		123	.070	.295	.060	.253	45N	86W

[b]74	5-10-77	11.9		Uncertain[f]	.034	.107	.094	.296	46N	92W
[b]75	5-11-77	11.9		[e]-12	.100	.315	.112	.353	48N	100W
[b]76	5-11-77	11.9		107	.106	.334	.162	.510	56N	137W
77	5-12-77	13.7	60	151	.072	.304	.149	.628	63N	152W
78	5-14-77	10.1	90	25	.127	.310	.202	.493		152W
79	5-14-77	10.7		49	.129	.340	.181	.477		152W
80	5-15-77	10.1		37	.154	.376	.193	.471		149W
81	5-15-77	8.8		[e]-23	.143	.300	.070	.147		152W
82	5-16-77	12.5		91	.065	.226	.095	.330	70N	147W
83	5-17-77	9.8		-43	.123	.289	.113	.266	63N	152W
84	5-17-77	12.8		61	.092	.336	.173	.632	63N	152W
[b]85	5-18-77	12.5		38	.085	.295	.202	.700	56N	137W
[b]86	5-19-77	11.3		[e]2	.090	.258	.156	.446	48N	100W
[b]87	5-19-77	12.5		[e]-8	.042	.146	.130	.451	46N	92W
88	7-26-77	12.8	82	20	.037	.135	.107	.391	45N	86W
89	7-26-77	12.8	90	20	.022	.080	.059	.215		
90	7-27-77	12.5		14	.035	.122	.088	.306		
91	7-28-77	13.4		55	.027	.109	.072	.289		
92	8-1-77	11.6		33	.027	.081	.072	.217		
93	8-2-77	12.5		14	.033	.115	.116	.404	42N	
94	8-9-77	13.7		-5	.023	.097	.062	.261	42N	87W
95	8-11-77	12.2		15	.041	.136	.093	.308	45N	86W
96	8-12-77	12.8	60	8	.023	.084	.080	.292	46N	86W
97	8-15-77	11.9	60	8	.021	.066	.074	.234	48N	87W
98	9-20-77	12.5	90	63	.048	.167	.085	.296	46N	86W
99	9-22-77	12.2	93	19	.027	.091	.072	.240	46N	86W
100	9-26-77	13.1	90	78	.047	.181	.092	.352	46N	86W
101	9-27-77	13.1	55	-46	.016	.062	.049	.187	42N	87W
102	10-17-77	10.7	90	44	.030	.078	.091	.240	42N	80W
103	10-18-77	9.4	105	30	.057	.128	.085	.192	45N	85W
104	10-19-77	10.1	90	44	.034	.084	.086	.209	44N	84W
[g]105	10-25-77	11.3	90	-27	023 [h].016	.067 [h].047	.066 [h].051	.190 [h].146	44N	84W
[g]106	11-1-77	13.1	90	56	.014 [h].010	.054 [h].037	.025 [h].018	.096 [h].070	44N	84W
[g]107	11-8-77	13.7	75	122	.016 [h].000	.067 [h].000	.035 [h].025	.148 [h].107	42N	80W

[a]On flights 32-41, 42-48, 54-63, 77-84, and 105-107, paired alternative filters were used for experimental comparisons. All other flights were with matched filters in each pod. Unless otherwise indicated, data are presented only for untreated IPC-1478 filter samples; data for matched samples were averaged.

[b]Transit flights.

[c]Sulfate analysis by spectrophotometric technique.

[d]Nitrate analysis by electrochemical technique.

[e]Average interval based on mixed sampling below and above tropopause.

[f]Uncertain tropopause location.

[g]Flight with comparison filter sampling: IPC-1478 and glass fiber.

[h]Glass-fiber filter data.

TABLE II. - FILTER DATA FROM GASP-747 AIRCRAFT - SINGLE-FILTER APPARATUS

Filter	Exposure date	Altitude, km	Exposure time, min	Region[a]	$SO_4^=$ concentration, $\mu g/m^3$	$SO_4^=$ mixing ratio, ppbn	NO_3^- concentration, $\mu g/m^3$	NO_3^- mixing ratio, ppbm	Average latitude, deg	Average longitude, deg
411	2-27-76	11.9	83	T	[b]0.059	[b]0.187	[c]0.070	[c]0.222	37.5 N	146.5 E
401	3-3-76	10.7,11.9	90,30	U	[b].012	[b].032	[c].026	[c].069	29. 5N	67.5 E
503	3-3-76	11.3,11.9	65,44	S	[b].185	[b].533	[c].098	[c].282	40.0 N	101.5W
508	3-9-76	10.7	120	U	[b].147	[b].389	[c].084	[c].222	56.0 N	14.0W
604	3-18-76	10.7	120	T	[b].038	[b].100	[c].063	[c].167	40.5 N	85.5W
712	7-15-76	11.3	120	U	.148	.426	.096	.276	52.0 N	21.0W
514	8-13-76	10.1	120	U	.067	.164	.072	.177	56.5 N	164.5W
422	9-2-76	10.7,11.3	14,72	T	.051	.147	.140	.403	11.5 N	74.5W
516	9-20-76	10.7,11.9	65,55	U	.053	.140	.060	.159	42.0 N	86.0W
423	11-24-76	11.0	119	T	.033	.091	.028	.077	34.0 N	133.5W
236	12-22-76	10.7,11.3	92,23	U,S	.051	.135	.055	.145	42. 5N	85.0W
333	12-28-76	11.3,11.9,12.5	40,50,30	T,T,S	.041	.130	.091	.288	42.0 N	109.5W
419	10-17-76	11.3	122	T	.029	.084	.051	.147	13.5 N	106.5 E
425	10-20-76	11.9	55	U	.020	.063	.055	.174	43.0 N	122.5W
335	12-10-76	11.0	120	U	.019	.052	.015	.041	33.5 N	134.5W
239	12-25-76	11.0	30	T	.026	.071	.002	.005	33.5 N	123.5W
434	12-31-76	10.7	121	T	.023	.061	.007	.018	29.5 N	67.5W
338	1-9-77	10.7	120	U	.030	.079	.032	.085	32.5 N	130.5W
430	1-3-77	11.0	120	U	.054	.148	.050	.137	34.2 N	132.8W

[a]S denotes stratosphere; T denotes troposphere; U denotes uncertain region.
[b]Sulfate analysis by spectrophotometric technique.
[c]Nitrate analysis by electrochemical technique.

TABLE III. - FILTER DATA FROM GASP-747 AIRCRAFT - MULTIFILTER APPARATUS

Filter	Exposure date	Altitude, km	Exposure time, min	Region[a]	$SO_4^=$ concentration, $\mu g/m^3$	$SO_4^=$ mixing ratio, ppbm	NO_3^- concentration, $\mu g/m^3$	NO_3^- mixing ratio, ppbm	Average latitude, deg	Average longitude, deg
201-2	8-1-77	11.0	120	T	0.023	0.064	0.049	0.135	31.0N	131.2W
201-3	8-4-77	11.0	120		.007	.021	.021	.057	34.2N	132.7W
201-4	8-7-77	11.6	120		.011	.034	.020	.062	25.4N	146.9W
201-5	8-10-77	10.4,11.6	30,90		.004	.013	.005	.014	24.8N	142.9W
201-6	8-13-77	11.2	61		.007	.020	.022	.063	37.2N	111.0W
201-8	8-16-77	11.0	120		.036	.104	.039	.110	33.6N	134.2W
401-1	7-8-77	11.3,10.5	10,6		.000	.000	.000	.000	35.8N	120.0W
401-2	7-11-77	11.3,9.7	50,6		.005	.016	.004	.011	18.3N	118.2E
401-3	7-17-77	11.3	51		.005	.014	.004	.013	18.3N	118.3E
401-4	7-20-77	10.7	14		.046	.123	.000	.000	35.7N	120.8W
401-5	7-23-77	10.1	120		.016	.039	.012	.031	17.4S	164.1E
401-6	7-26-77	10.7,11.3	100,20		.003	.008	.002	.006	7.0S	174.4E
401-7	7-29-77	10.7	120		.006	.016	.012	.031	38.6N	39.4E
401-8	8-13-77	11.3	120		.018	.053	.053	.154	12.6N	78.8W
501-5	9-3-77	10.1	120		.041	.101	.029	.071	50.7N	100.3W
501-6	9-6-77	10.7	120		.043	.114	.016	.042	45.0N	57.8W
501-7	9-11-77	10.1	77		.006	.015	.021	.053	29.3N	61.0E
501-8	9-15-77	10.1	120		.045	.110	.032	.078	47.8N	57.6W
301-1	4-15-77	10.7	120		.012	.031	.019	.051	32.8N	133.6W
301-3	4-21-77	10.7,11.9	75,45		.018	.051	.025	.071	39.7N	86.1W
301-4	4-24-77	11.0	120	S	.055	.150	.042	.114	31.9N	129.4W
202-1	10-6-77	10.1	120	T	.018	.044	.008	.021	46.7N	145.9W
202-2	10-9-77	11.9	113		.014	.046	.009	.027	30.4N	58.6E
202-3	10-12-77	11.3	120		.009	.026	.016	.046	22.9N	87.2E
202-5	10-15-77	10.1			.018	.044	.057	.140	14.4N	107.4E
202-6	10-18-77	10.1		U	.047	.116	.031	.077	46.4N	58.1W
202-7	10-21-77	10.7		U	.022	.059	.052	.139	30.6N	131.0E
202-8	10-24-77	10.7,11.3	90,30	S	.061	.164	.046	.126	44.8N	59.0W
402-4	11-11-77	11.3	120	T	.020	.058	.014	.042	40.2N	108.2W
402-5	11-14-77	11.9	120	T	.037	.117	.031	.099	30.4N	130.3W
502-5	1-9-78	10.1	115	S	.084	.207	.054	.133	53.3N	154.8W
801-8	2-17-78	10.8	120	S	.063	.169	.072	.194	42.2N	85.7W
303-2	1-6-78	11.1	89	T	.001	.006	.000	.000	35.2N	130.4W
303-3	1-9-78	10.1	18	S	.041	.100	.169	.415	35.9N	120.5W
303-4	1-15-78	11.3	47	S	.034	.098	.057	.164	41.6N	80.4W
303-5	1-18-78	12.2	51	T	.019	.064	.000	.000	23.6N	151.5W
303-6	1-21-78	11.3	115		.036	.104	.061	.177	37.4N	107.3W
303-8	1-24-78	10.4,11.6	79,41		.022	.059	.042	.113	27.2N	148.6W
701-2	3-7-78	11.0	115		.008	.023	.009	.026	34.4N	132.6W
701-3	3-10-78	11.0	55		.015	.040	.016	.043	35.8N	128.7W
701-4	3-12-78	11.3	40		.018	.050	.037	.106	25.4N	154.4W
701-6	3-16-78	10.1,11.3	30,80		.010	.032	.012	.035	25.4N	144.5W
701-8	3-20-78	11.9,9.5	26,4		.007	.021	.022	.065	23.6N	155.8W
304-2	4-18-78	10.3,11.9	15,105	S	.060	.185	.147	.455	39.6N	94.5W
304-3	4-21-78	11.0	120	T	.021	.059	.033	.090	34.1N	133.2W
304-4	4-24-78	11.3	120	S	.044	.126	.096	.277	40.2N	108.2W
304-5	4-27-78	11.3	110	T	.011	.032	.053	.153	26.6N	147.8W
304-6	4-28-78	11.0	105	T	.035	.095	.058	.159	34.4N	132.4W
304-8	5-3-78	11.3,10.7	40,10	S	.040	.112	.135	.381	36.2N	111.6W
802-8	5-18-78	10.1,10.7	70,50	T	.065	.165	.123	.311	32.0N	149.3W

[a]S denotes stratosphere; T denotes troposphere; and U denotes uncertain region.

TABLE III. - Concluded.

Filter	Exposure date	Altitude, km	Exposure time, min	Region[a]	$SO_4^=$ concentration, $\mu g/m^3$	$SO_4^=$ mixing ratio, ppbm	NO_3^- concentration, $\mu g/m^3$	NO_3^- mixing ratio, ppbm	Average latitude, deg	Average longitude, deg
702-2	5-18-78	11.6	120	T	0.018	0.054	0.035	0.105	24.7N	143.3W
702-4	5-24-78	11.3	110	S	.048	.147	.072	.222	40.2N	108.8W
702-5	5-27-78	11.3,11.9,11.3	97,5,19	S	.055	.158	.062	.178	40.3N	108.6W
702-8	5-30-78	11.3	120	T	.034	.103	.031	.094	27.1N	146.7W
305-2	6-8-78	11.3	120		.014	.039	.012	.034	25.4N	144.2W
305-3	6-11-78	11.9	120		.010	.033	.012	.036	40.6N	86.4W
305-4	6-14-78	10.7,11.9	54,66		.046	.133	.086	.251	41.2N	86.4W
305-5	6-17-78	10.7	60	S	.091	.240	.130	.343	36.4N	129.8W
305-8	6-20-78	11.9,9.6	30,11	U,T	.021	.661	.089	.257	40.8N	80.8W
103-4	6-17-78	10.1	121	T	.010	.025	.033	.081	27.2N	124.4E
203-2	7-29-78	10.1	120	S	.096	.234	.083	.202	52.4N	20.0W
203-4	8-1-78	10.7,11.3	50,70	T	.020	.063	.044	.135	36.2N	84.2W
203-5	8-4-78	9.8	120		.022	.053	.021	.049	31.1N	130.8W
203-7	8-10-78	10.7	55		.009	.024	.020	.053	17.6S	46.4W
203-8	8-13-78	10.4	120						27.8N	147.3W
203-8	8-16-78	10.1	120	U	.032	.079	.027	.066	51.0N	103.8W
306-3	10-3-78	10.3	120	T	.071	.180	.023	.058	52.4N	34.7W
306-6	10-6-78	9.8	120		.029	.069	.036	.086	51.7N	22.6W
104-4	9-27-78	11.3	120		.004	.012	.010	.030	25.7N	143.6W
104-6	10-3-78	11.3	77		.014	.040	.034	.098	25.1N	150.6W
803-3	10-12-78	10.1	75		.030	.074	.025	.062	53.2N	15.2W
803-6	10-15-78	10.7	120		.006	.015	.003	.008	30.8N	132.1E
803-8	10-21-78	10.1	120		.046	.115	.044	.108	54.2N	103.6W
704-3	10-6-78	11.3	120		.003	.008	.010	.030	28.3N	144.4W
704-4	10-6-78	11.3	60		.015	.043	.015	.044	33.8N	132.0W
704-5	10-6-78	11.3	18		.000	.000	.023	.066	35.9N	125.8W
704-6	10-9-78	11.9,12.5	80,40	U	.010	.032	.037	.121	34.9N	105.0W
704-8	10-12-78	11.2	50	T	.000	.000	.050	.141	35.2N	128.3W
105-2	11-8-78	11.0	120		.006	.016	.007	.020	33.6N	144.4W
105-3	11-11-78	10.7,11.9	30,90		.010	.032	.004	.012	39.6N	86.0W
105-6	12-5-78	11.3	60		.010	.030	.009	.027	38.0N	117.8W
105-8	12-5-78	11.3			.015	.043	.019	.056	40.6	105.3W
307-2	10-27-78	11.9			.010	.032	.052	.165	41.3N	86.0W
307-3	10-27-78				.017	.053	.056	.178	41.2N	96.0W
307-4	10-27-78				.012	.040	.065	.207	40.0N	105.5W
307-5	10-27-78		55		.014	.043	.032	.102	38.5N	115.6W
307-6	10-30-78	11.2	60		.010	.030	.021	.060	27.0N	147.5W
307-8	10-30-78	11.2	60		.013	.037	.027	.078	31.2N	138.6W
705-6	12-17-78	11.0	120	U	.019	.051	.058	.159	34.4N	132.3W
903-2	12-23-78	11.3	51	T	.007	.021	.044	.129	34.9N	119.9W
903-3	12-26-78	11.6	60	T	.010	.030	.028	.084	27.9N	147.6W
903-5	12-29-78	11.3	60	S	.027	.078	.067	.192	35.7N	117.0W
903-6	12-29-78	11.3	60	U	.022	.062	.059	.169	38.9N	105.7W
903-8	12-29-78	11.3	43	S	.016	.046	.079	.228	40.8N	94.6W

[a]S denotes stratosphere; T denotes troposphere; and U denotes uncertain region.

TABLE IV. - AVERAGED FILTER DATA[a]

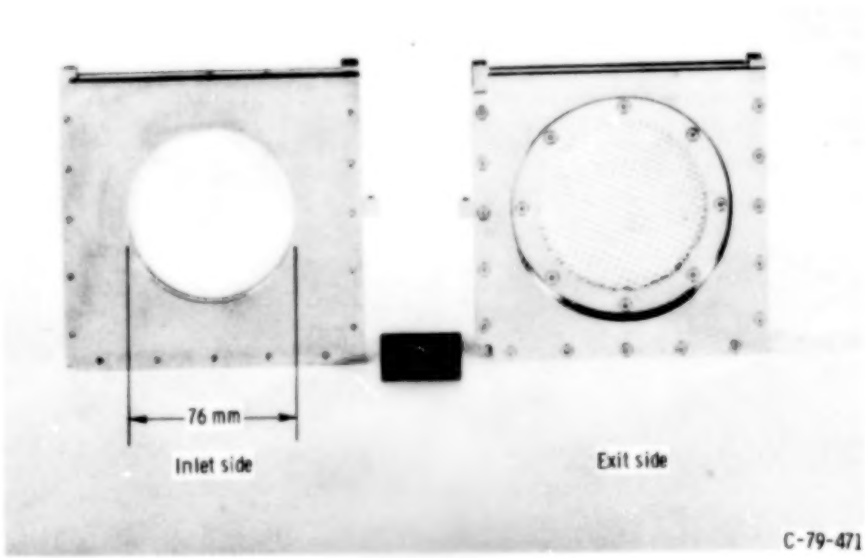
Data group	Aircraft data source	Seasonal group	Region[b]	Altitude range, km	Number of samples, n	$SO_4^=$ mixing ratio, ppbm		Number of samples, n	NO_3^- mixing ratio, ppbm	
						Mean, $\bar{X}$	Standard deviation, S		Mean, $\bar{X}$	Standard deviation, S
A	F-106	Winter-spring	S	10.1-13.7	30	0.349	0.105	30	0.492	0.202
		Summer-fall	S	9.4-13.4	19	.109	.036	19	.263	.076
		Winter-spring	T	9.8-12.5	3	.162	-----	3	.110	-----
		Summer-fall	T	11.3-13.7	9	.089	.019	9	.176	.063
B	747	Winter-spring	S	10.1-11.9	13	0.173	0.116	13	0.243	0.097
		Summer-fall	S	10.1-11.3	2	.199	-----	2	.164	-----
		Winter-spring	T	9.5-12.2	25	.064	.045	25	.097	.080
		Summer-fall	T	9.7-11.9	48	.051	.039	48	.078	.069
C	747 and selected F-106	Winter-spring	S	10.1-12.5	29	0.258	0.134	29	0.354	0.174
		Summer-fall	S	9.4-12.5	14	.126	.044	14	.254	.067
		Winter-spring	T	9.5-12.5	28	.075	.060	28	.099	.085
		Summer-fall	T	9.7-12.5	52	.054	.039	52	.085	.073
D	747 and all F-106	Winter-spring	S	10.1-13.7	43	0.296	0.135	43	0.417	0.210
		Summer-fall	S	9.4-13.7	21	.118	.044	21	.254	.079
		Winter-spring	T	9.5-12.5	28	.075	.060	28	.099	.085
		Summer-fall	T	9.7-13.7	57	.057	.039	57	.093	.077

[a]Data from filters with exposure times less than 30 minutes, with uncertain identification of exposure region, or with mixed tropospheric and stratospheric exposure were excluded from these averages.

[b]S denotes stratosphere; T denotes troposphere.

C-73-2675

(a) Internal view of single-filter mechanism; filter holder at midposition.



C-79-471

(b) Filter-holder assembly for single-filter system.

Figure 1. - Single-filter system.

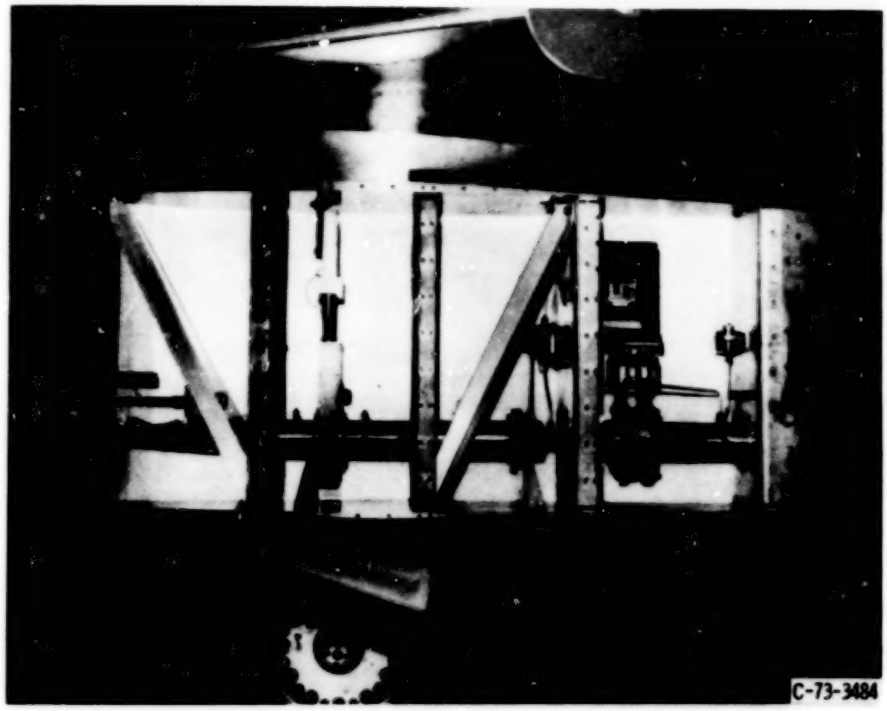


Figure 2. - Filter sampling system used on NASA F-106 aircraft.

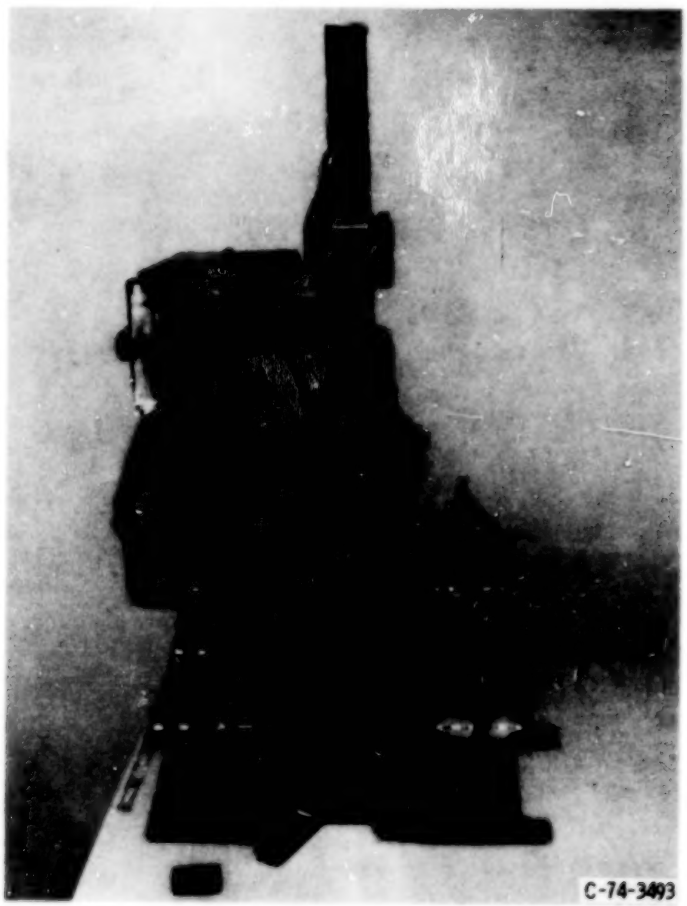


Figure 3. - Single-filter system modified for use on GASP 747 aircraft.

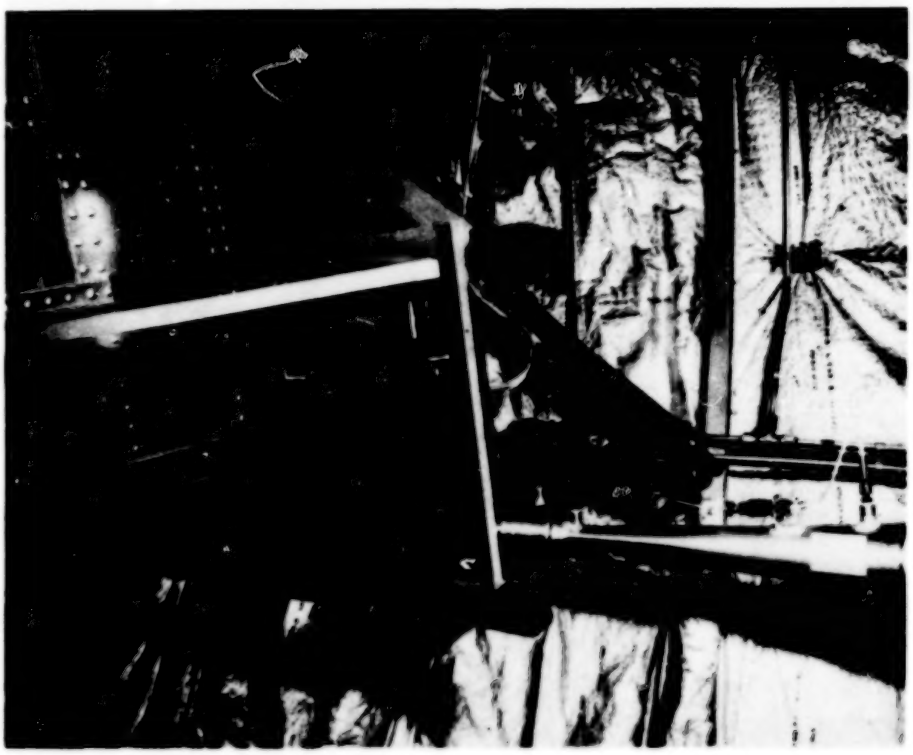
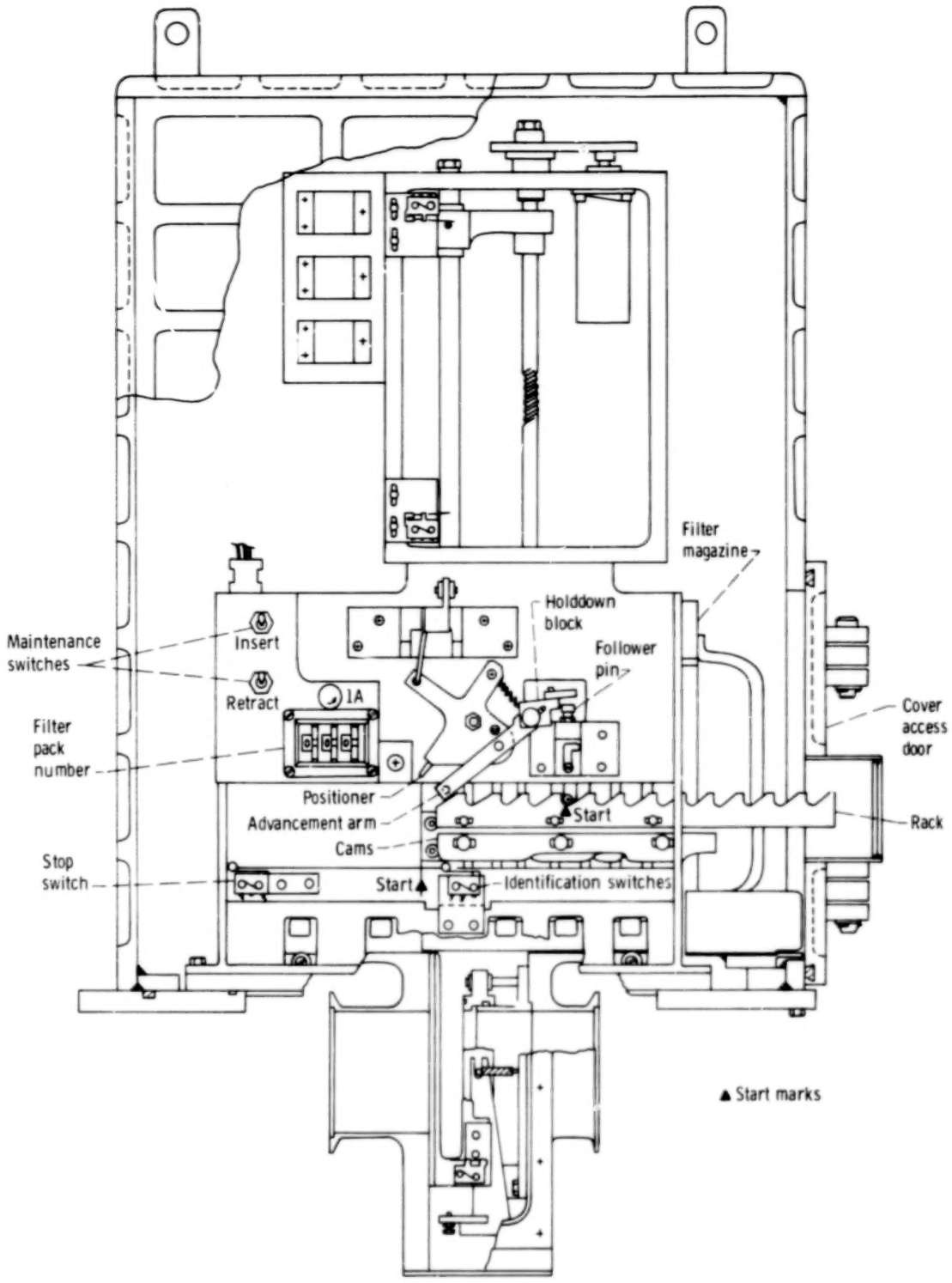
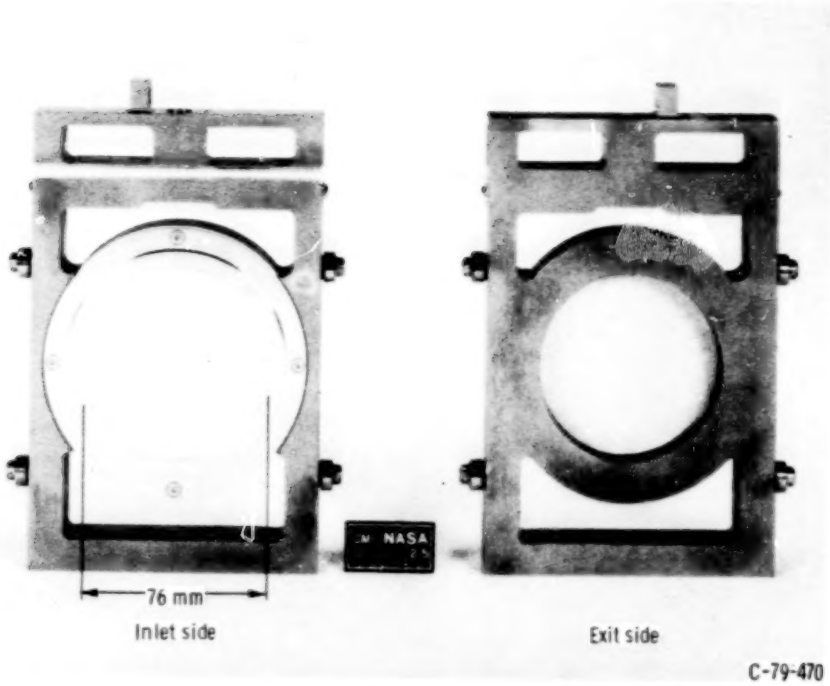


Figure 4. - Single-filter system installed on GASP 747 aircraft.

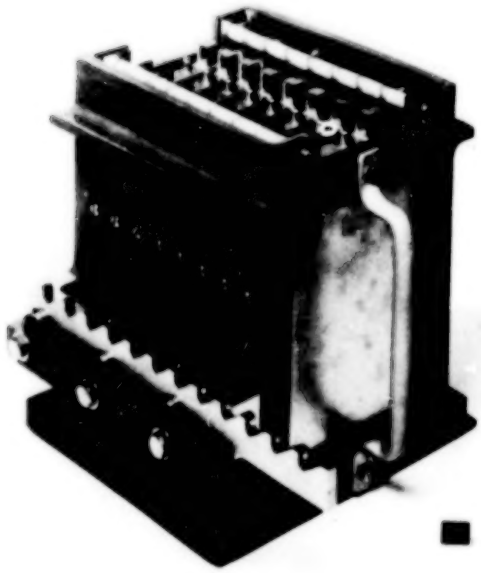


(a) Internal view of multifilter slide mechanism.

Figure 5. - Multifilter system.



(b) Filter-holder assembly for multifilter system.



(c) Magazine assembly for multifilter system.

Figure 5. - Concluded.

Figure 6. - Multifilter system installed on GASP 747 aircraft.

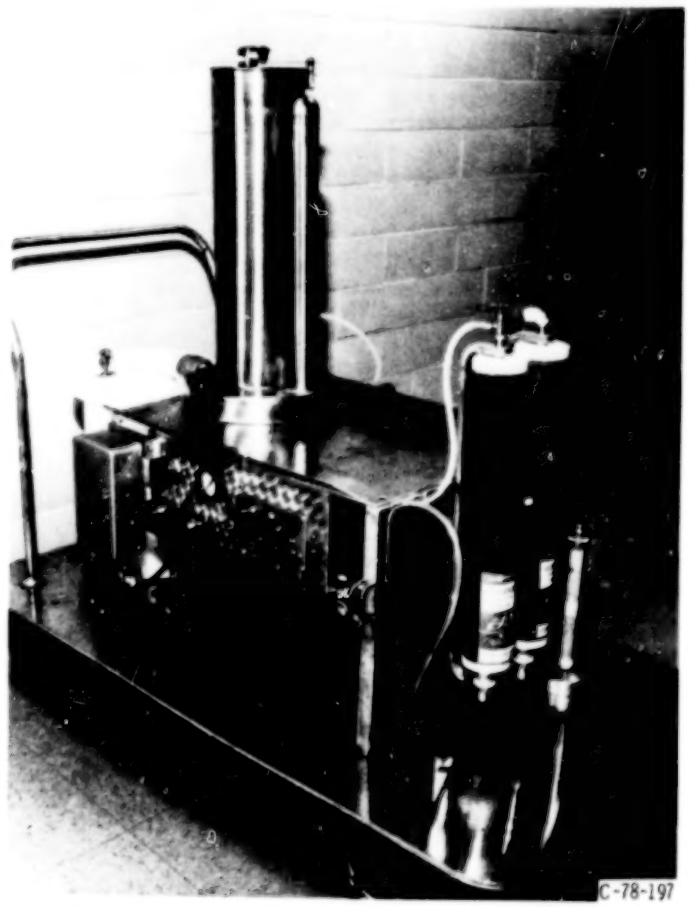


Figure 7. - Semiautomatic washing apparatus.

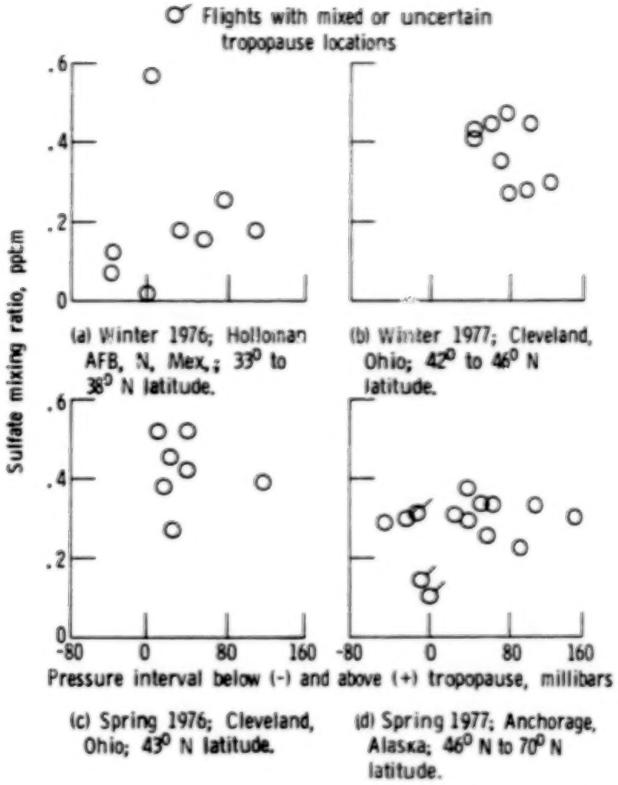

Flights with mixed or uncertain tropopause locations
Sulfate mixing ratio, ppbm
.6
.4
.2
0
-80
0
80
160
(a) Winter 1976; Holloman AFB, N. Mex.; 33° to 38° N latitude.
(b) Winter 1977; Cleveland, Ohio; 42° to 46° N latitude.
(c) Spring 1976; Cleveland, Ohio; 43° N latitude.
(d) Spring 1977; Anchorage, Alaska; 46° N to 70° N latitude.
Pressure interval below (-) and above (+) tropopause, millibars

Figure 8. - Winter-spring sulfate mixing ratios from F-106 flights near the tropopause.

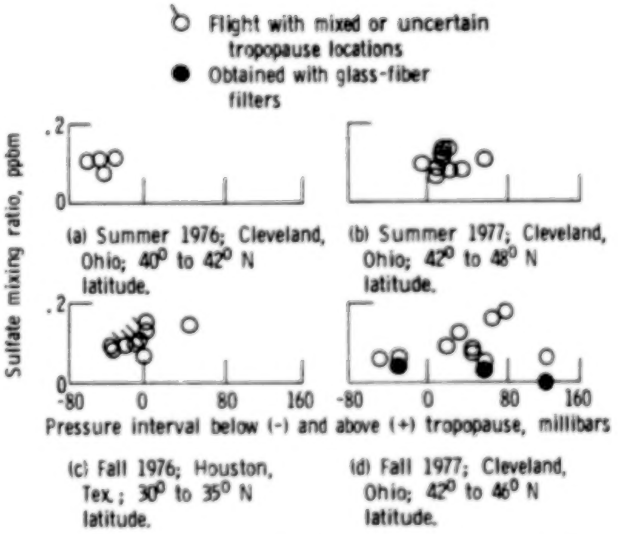
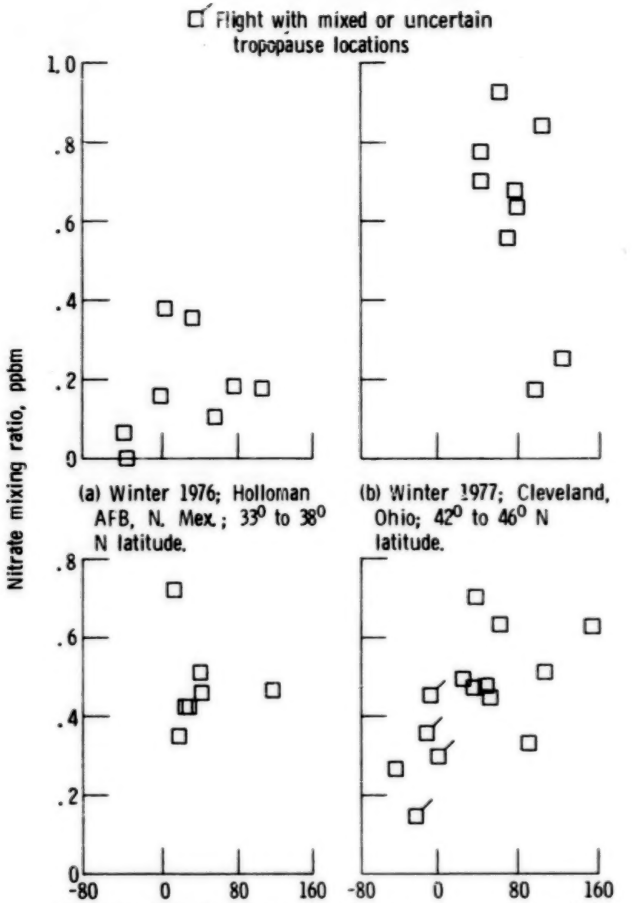

Flight with mixed or uncertain tropopause locations
Obtained with glass-fiber filters
Sulfate mixing ratio, ppbm
.2
0
-80
0
80
160
(a) Summer 1976; Cleveland, Ohio; 40° to 42° N latitude.
(b) Summer 1977; Cleveland, Ohio; 42° to 48° N latitude.
(c) Fall 1976; Houston, Tex.; 30° to 35° N latitude.
(d) Fall 1977; Cleveland, Ohio; 42° to 46° N latitude.
Pressure interval below (-) and above (+) tropopause, millibars

Figure 9. - Summer-fall sulfate mixing ratios from F-106 flights near the tropopause.



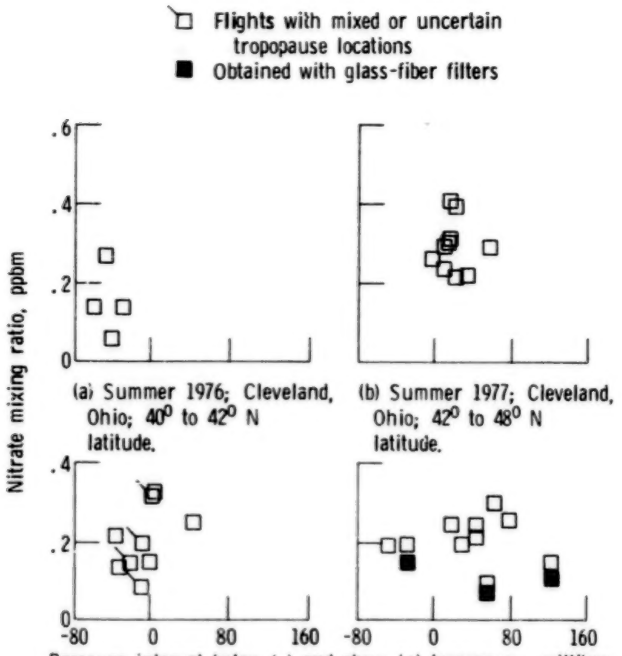
(a) Winter 1976; Holloman AFB, N. Mex.; 33° to 38° N latitude.

(b) Winter 1977; Cleveland, Ohio; 42° to 46° N latitude.

(c) Spring 1976; Cleveland, Ohio; 43° N latitude.

(d) Spring 1977; Anchorage, Alaska; 46° N to 70° N latitude.

Figure 10. - Winter-spring nitrate mixing ratios from F-106 flights near the tropopause.



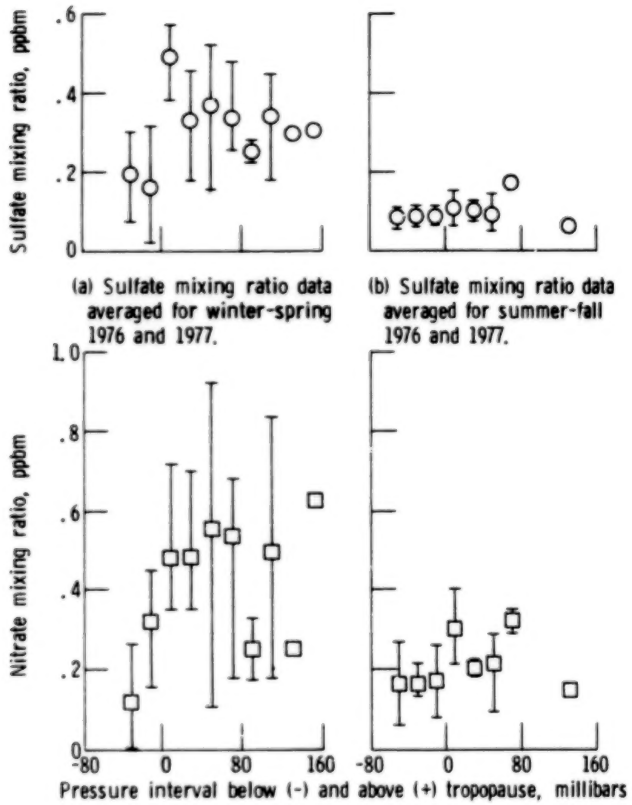
(a) Summer 1976; Cleveland, Ohio; 40° to 42° N latitude.

(b) Summer 1977; Cleveland, Ohio; 42° to 48° N latitude.

(c) Fall 1976; Houston, Tex.; 30° to 35° N latitude.

(d) Fall 1977; Cleveland, Ohio; 42° to 46° N latitude.

Figure 11. - Summer-fall nitrate mixing ratios from F-106 flights near the tropopause.



(a) Sulfate mixing ratio data averaged for winter-spring 1976 and 1977.

(b) Sulfate mixing ratio data averaged for summer-fall 1976 and 1977.

(c) Nitrate mixing ratio data averaged for winter-spring 1976 and 1977.

(d) Nitrate mixing ratio data averaged for summer-fall 1976 and 1977.

Figure 12. - Averaged sulfate and nitrate mixing ratio data from F-106 flights near the tropopause. The range of data included within each 20-millibar pressure interval is as indicated by the vertical bars.

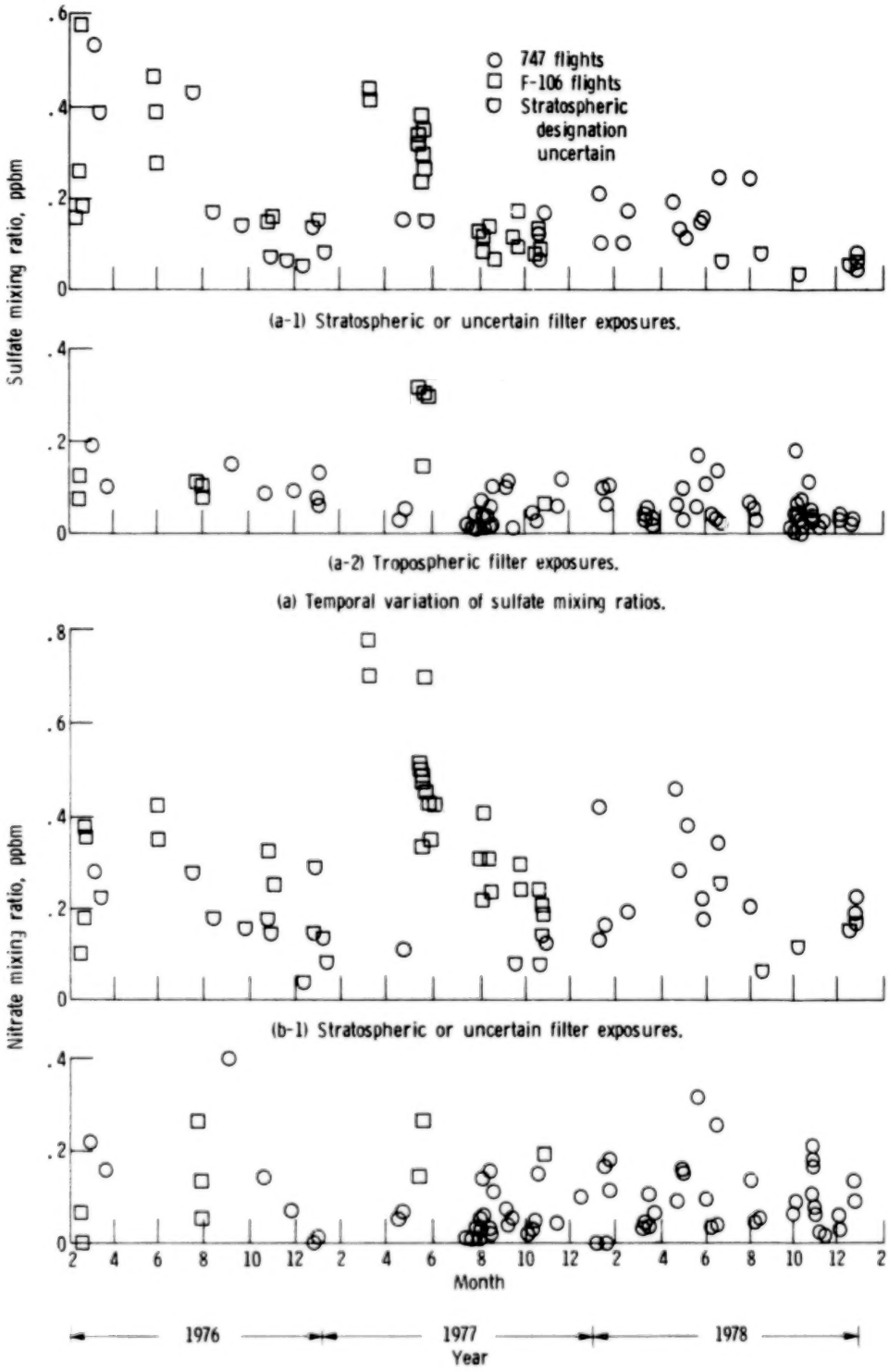


Figure 13. - Combined filter data from Boeing 747 and F-106 aircraft in chronological order for altitudes at or below 12.5 kilometers.

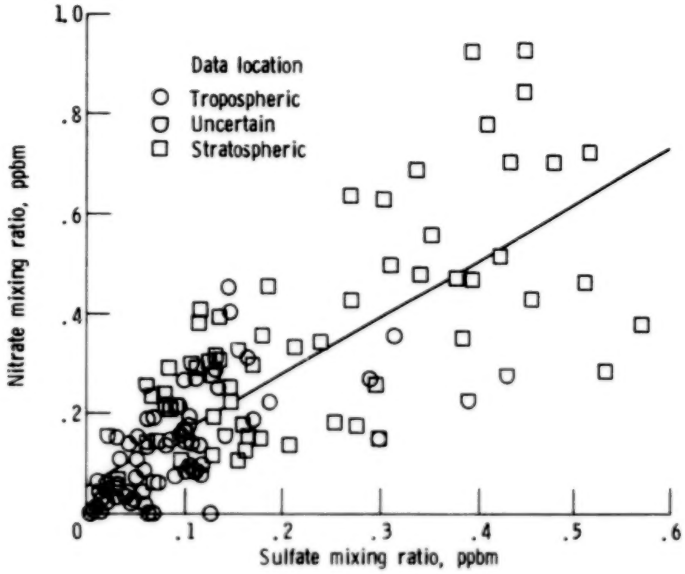


Figure 14. - Correlation of nitrate and sulfate mixing ratios. Correlation coefficient, 0.78; slope, 1.12; intercept, 0.052.

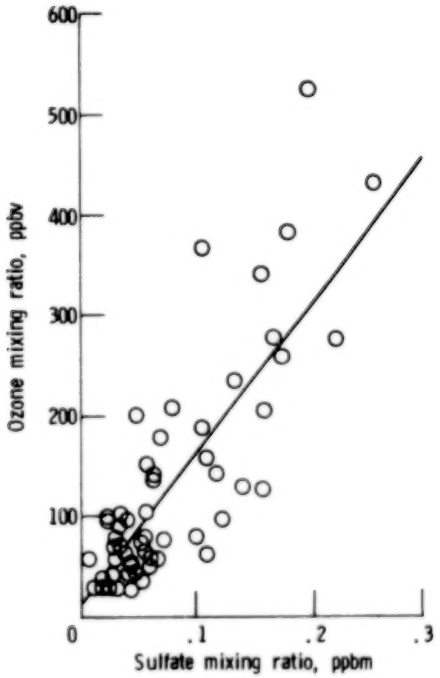


Figure 15. - Correlation of ozone and sulfate mixing ratios. Correlation coefficient, 0.80; slope, 1574; intercept, 15; number of samples, 62.

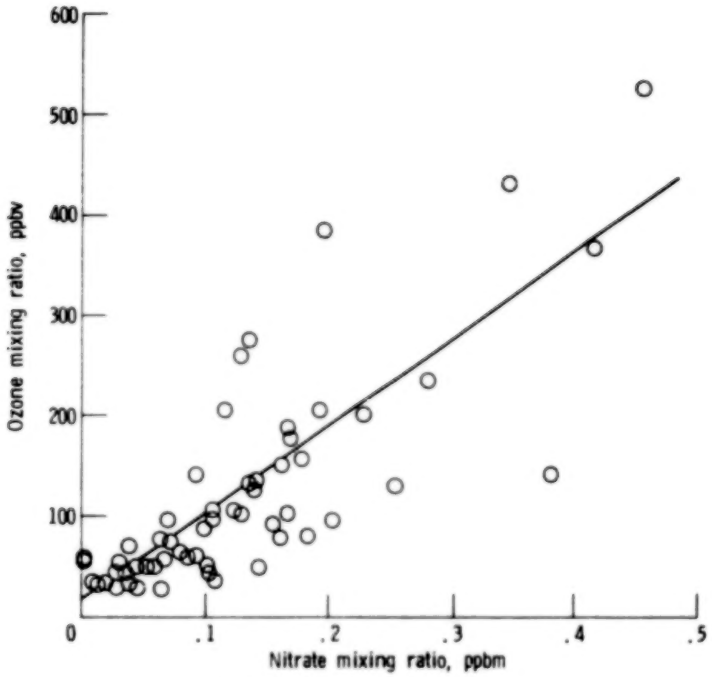


Figure 16. - Correlation of ozone and nitrate mixing ratios. Correlation coefficient, 0.79; slope, 868; intercept, 17; number of samples, 62.

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15. Supplementary Notes

16. Abstract

Filter samples collected near the tropopause during 1976, 1977, and 1978 with a NASA Lewis F-106 aircraft and two commercial Boeing 747 aircraft were analyzed for sulfate and nitrate ion content. These sampling platforms were operated as part of the NASA Global Atmospheric Sampling Program (GASP). Within the range of routine commercial flight altitudes (at or below 12.5 km), stratospheric mass mixing ratios averaged 0.26 ppbm for sulfate and 0.35 ppbm for nitrate for the winter-spring group and 0.13 ppbm for sulfate and 0.25 ppbm for nitrate for the summer-fall group. Tropospheric mass mixing ratios averaged 0.08 ppbm for sulfate and 0.10 ppbm for nitrate for the winter-spring group and 0.05 ppbm for sulfate and 0.08 ppbm for nitrate for the summer-fall group. Correlations of the filter data with available ozone data suggest that the sulfate and nitrate are transported from the stratosphere to the troposphere.

17. Key Words (Suggested by Author(s))	18. Distribution Statement
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